

Dispersion and damping of multi-quantum well polaritons from resonant Brillouin scattering by folded acoustic modes

B. Jusserand,¹ A. Fainstein,² R. Ferreira,³ S. Majrab,¹ and A. Lemaitre⁴

¹*Institut des Nanosciences de Paris, CNRS UMR7588, UPMC, 75005 Paris, France*

²*Instituto Balseiro and Centro atomico Bariloche, CNEA, R8402AGP Bariloche, Argentina*

³*Laboratoire Pierre Aigrain, ENS, CNRS UMR 8551, UPMC, UPD, 75005 Paris, France*

⁴*Laboratoire de Photonique et de Nanostructures, CNRS UPR20, 91460 Marcoussis, France*

We report on confined exciton resonances of acoustic and folded acoustic phonon light scattering in a GaAs/AlAs multi-quantum-well. Significant variations of the line shifts and widths are observed across the resonance and quantitatively reproduced in terms of the polariton dispersion. This high resolution Brillouin study brings new unexpectedly detailed informations on the polariton dynamics in confined systems.

Resonant Brillouin scattering has played a very important role in the experimental validation of the concept of exciton polariton in bulk semiconductors and has provided the first determination of the specific energy dispersion of these excitations close to the exciton transition [1]. In such a case, strong dispersion effects appear in the dielectric constant, significantly affecting the relation between the energy and the wavevector of the electromagnetic excitations. Based on the linear dispersion of acoustic waves in solids, a direct relation exists between the photon energy shift measured by Brillouin scattering and the dispersion in the compound of the incoming and outgoing polaritons involved in the scattering event. Strong variations of the Brillouin shift are then expected from the wavevector and energy conservation in the presence of dispersion. These effects have been reported in several bulk semiconductors and the energy variation was used to determine the polariton dispersion close to the fundamental exciton transition. When the finite lifetime of the exciton is taken into account, the dielectric constant becomes a complex quantity in the vicinity of the exciton energy and polariton damping is expected to emerge. The related variation of the Brillouin linewidth has escaped up to now the observation because of the limited resolution of standard Raman setups. Additional complications appear in the polariton picture in bulk materials because of the exciton energy dispersion: two polariton branches coexist at the same energy in some cases and the conversion of a single photon outside the sample into these multiple branches has never been completely understood [2]. More recently, the concept of exciton polariton has been extensively reconsidered in the context of microcavities [3]. In these photonic monolithic resonators, two-dimensional cavity photons with dispersion in the plane of the cavity strongly interact with two-dimensional excitons confined in quantum wells inserted in the photonic cavity, leading to a large number of novel optoelectronic properties. Inelastic light scattering has been applied to demonstrate the relevance of the polaritonic picture to describe light scattering by cavity polaritons [4]. It has not been possible however to follow the light scattering

spectra across the full excitonic resonance contrary to what was previously done for bulk polaritons. Moreover Brillouin studies with a determination of incoming energy resolved energy shifts are not available up to now. We will show in this letter that these issues can be conclusively addressed using high resolution Brillouin studies of high quality multi-quantum wells.

Surprisingly, semiconductor superlattices have attracted very little attention from the point of view of Brillouin scattering close to strong excitonic resonances, while being extensively studied for their unique optical properties. Moreover, superlattices exhibit acoustic phonon folding [5] due to new periodicity associated with the periodic alternance of semiconductors with contrasting acoustic and acousto-optic properties. This new feature opens the route for the investigation of exciton Brillouin resonances using several phonon probes with different energies, instead of the unique low energy Brillouin active acoustic phonon in bulk materials. A series of reports has been focused on the emergence at strong excitonic resonances of dominant contributions to the Brillouin signal from isolated quantum wells due to fluctuations of well widths from well to well [6]. The activation of the full density of acoustic states results from the localization of the intermediate states involved in the scattering process, while no evidence is obtained of polariton related resonant features on wavevector conserving acoustic phonons.

In multi-quantum wells, a type of superlattices in which the barriers are sufficiently thick to make negligible the coupling between electron states confined in neighboring quantum wells, excitons no longer display any significant dispersion along the growth axis [7]. This provides a unique realization of the simple theoretical picture [8] of a discrete level in strong interaction with a photonic continuum in experimental situations where the in plane wavevector is fixed to zero by the geometry. In this case and taking into account excitonic damping, the polaritonic and the excitonic pictures merge together into a common description with a single excitation branch with significant dispersion and damping close to

the excitonic energy. This excitation is well described as a photon dressed by a dispersive complex dielectric constant.

We report in this letter Brillouin scattering by acoustic (LA) and folded acoustic (FLA) longitudinal modes in a GaAs/AlAs multi-quantum well. We have been able to follow the variations across the excitonic resonances of the energy, the width and the intensity of several well defined acoustic branches showing that a well defined phonon wavevector is preserved throughout the whole energy range. Thanks to the highest energy of the folded phonons, dispersion of polariton energy and damping in the incoming and the outgoing channels are well separated while there are mixed when the low energy Brillouin line is considered. Consistent informations on the background dielectric constant, on the excitonic oscillator strength and damping are extracted from the different experimental observations on LA and FLA modes.

The eigenvalue problem for interacting photons and excitons has been first solved for bulk materials by Hopfield [8] introducing new coupled excitations called exciton polaritons. The polariton eigenvalue equation writes:

$$\frac{c^2 k^2}{\omega^2} = \varepsilon_0 + \frac{4\pi\beta\omega_X^2}{\omega_X^2 - \omega^2}$$

in which $\hbar ck$ is the photon energy and $\hbar\omega_X$ the exciton one. ε_0 is the background dielectric constant and β a number describing the strength of the exciton-photon coupling. This equation leads to the existence of two branches of polaritons, the lower branch with energies extending from zero to $\hbar\omega_X$ and the upper branch extending from $\hbar\omega_X\sqrt{1 + \frac{4\pi\beta}{\varepsilon_0}}$ to infinity. Introducing $Q = \frac{4\pi\beta}{\varepsilon_0}$, the new dispersion can be described by two parameters: a) the longitudinal transverse splitting ($\omega_{LT} \simeq Q\omega_X/2$ for small values of β), the width of the gap between the lower and the upper polariton branches in which no states exist due to the polariton coupling and b) the Rabi splitting ($\omega_R \simeq \omega_X\sqrt{Q}$), the minimum separation between the two branches at a given wavevector. The weight of the photon and exciton components in the polariton wavefunctions strongly varies around the exciton energy in a range given by ω_R . Using standard values of the polariton dispersion in GaAs [1], the dispersion parameters read: $\omega_{LT} = 0.086\text{meV}$ and $\omega_R = 16\text{meV}$. Brillouin scattering is strongly affected by the polaritonic coupling. In the standard picture, the incoming photon is scattered into an outgoing one with the emission of an acoustic phonon whose energy and wavevector is obtained from the energy and wavevector conservation during the scattering process. As sound velocities are much smaller than light velocities in solids, the phonon wavevector equals twice the incoming one in the usual backscattering configuration, to an excellent approximation. In the presence of polaritons, two incoming and two outgoing branches have to be considered and acoustic phonons with up to

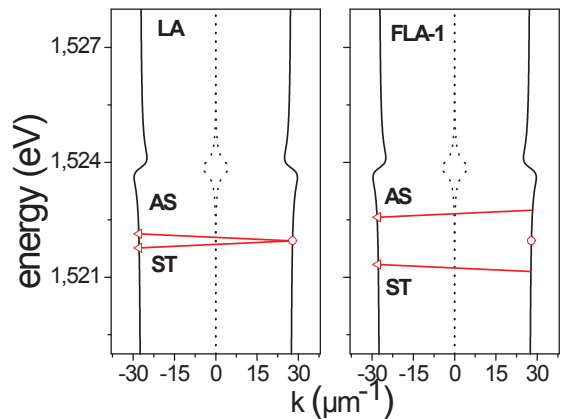


FIG. 1: Schematic description of the resonant Brillouin process between an ingoing polariton (red circles) and outgoing polaritons (red triangles) for Stokes (ST) and anti-Stokes (AS) processes with the LA phonon (left panel) and the lowest folded phonon FLA-1. The solid lines (resp. dashed lines) show the real part (resp. the imaginary part) of the polariton wavevector deduced from Eq.(1). The parameters of the calculations are typical for the studied sample.

four different wavevectors can be simultaneously involved in the scattering process [1]. Moreover, the wavevectors and thus the Brillouin shifts directly reflect the polariton dispersion. Experimental observations of such features are considered as one of the most direct evidence of the existence of excitonic polaritons in bulk semiconductors.

It has been demonstrated [7] that the previous analysis can be extended, using an effective β , to superlattices in the long wavelength limit, which applies to the optical properties studied here. Moreover a more realistic expression for the polariton is obtained when the homogeneous broadening of the exciton due to its finite lifetime is included:

$$\frac{c^2 k^2}{\omega^2} = \varepsilon_0 + \frac{4\pi\beta\omega_X^2}{\omega_X^2 - \omega^2 - i\gamma\omega} \quad (1)$$

The combination of damping and dispersion in the exciton part of the polariton leads to a complex situation which has not been described consistently as regards its consequences on Brillouin scattering. However the case of a discrete non dispersive exciton appears to be a simple model system whose experimental realization has never been considered up to now. In this case, a single polariton branch exists at any energy and its dispersion and damping can be described with a dispersive complex dielectric constant $\varepsilon(\omega)$ equal to the right hand side of Eq.(1). We illustrate in Fig. 1 the Brillouin scattering mechanism in this situation for two cases: in the left panel, the low energy Brillouin line LA with dispersion $\omega = vk$ (v is the sound velocity) is involved and the incoming and

outgoing polaritons take place at very close points in the dispersion. When the energy is in close resonance with the exciton, both the incoming and the outgoing channels contribute simultaneously to the resonance. In the right panel, the lowest folded acoustic line FLA-1 with dispersion $\omega = v(\frac{2\pi}{d} - k)$ (d is the period of the superlattice) is involved and the constant energy offset due to folding gives a unique possibility to separate the incoming and outgoing photon contributions. In this figure we used a damping of 0.3 meV, a value corresponding to the experiments discussed below. This damping is significantly larger than the longitudinal transverse splitting, thus letting the LT splitting irrelevant, but much smaller than the Rabi gap, leading to significant modification of the dielectric constant around the exciton energy due to the polariton coupling, which is the topics of the experimental study presented in this letter.

We have performed resonant Brillouin scattering on a multi-quantum well with 40 periods with 17.1 nm of GaAs and 7.5 nm of AlAs in each period. The Brillouin experiments have been performed with a Coherent MBR single-mode Ti-Sa tunable ring laser with wavelength stability better than 0.01 cm^{-1} . Resonance profiles have been measured close to different confined exciton levels and we focus here on the E1-HH1 transition between the lowest conduction electron state and the one of the heavy hole. The resonance curve has been followed using laser energy steps close to $25 \mu\text{eV}$ (0.2 cm^{-1}). The light emitted by the sample, including excitonic luminescence, Brillouin scattering and the peak at the laser energy, has been dispersed by a Dilor XY triple Raman spectrometer in additive mode and recorded with a nitrogen cooled CCD multichannel detector. Experimental resolution is close to 0.2 cm^{-1} . We discuss in this letter experimental results obtained at 80K. At higher temperatures, all intensities decrease, thus reducing the accuracy of the measurements. At lower temperature, the excitonic luminescence increases strongly and dominates over the Brillouin spectra below 20K. Thanks to the large intensity of the Brillouin lines in the intermediate temperature range (50-80K), we have been able to record in the same CCD window the different scattered lines without having to filter out the laser line or the exciton luminescence. This allows a very accurate determination of the energy shifts as a function of the incident laser energy.

We show in Figure 2 Brillouin spectra measured at a few different incident energies across the excitonic transition. The Brillouin line, with a shift close to 1.3 cm^{-1} , and the lowest folded lines, with shifts around 5 and 8 cm^{-1} , are recorded on each side of the laser line, respectively corresponding to Stokes and anti-Stokes processes, and on the top of the excitonic luminescence. In the left panel, one clearly sees the large variations of the intensity, plotted in logarithmic scale, and of the linewidth of the different peaks. In the spectrum in green, with laser

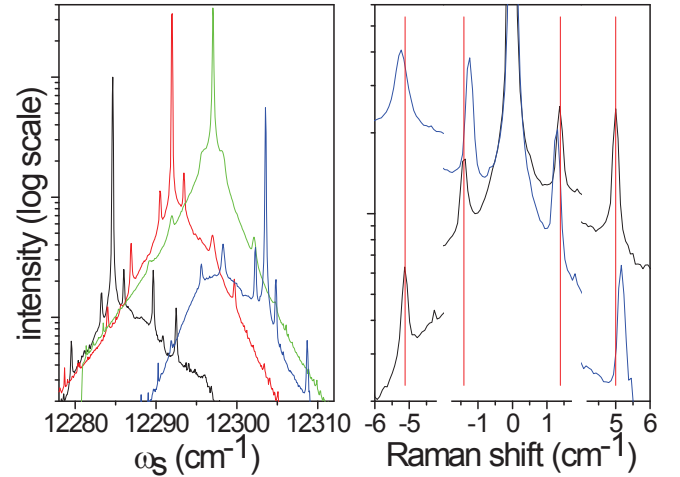


FIG. 2: Left panel: resonant Brillouin spectra at four different incident laser energies across the E1HH1 excitonic resonance. In the right panel, the two curves with the smallest and the largest incident energies in the left panel are shown with an energy scale relative to the laser position (Raman shift). The vertical lines point out the change in the Raman shifts across the resonance.

energy close to the exciton transition energy, all peaks are significantly broadened. In the spectra in blue and red, the folded lines with absolute energies close to the exciton transition are significantly broadened as well. When plotted against the laser energy (right panel), a variation of the line shifts becomes apparent, going towards opposite directions for the Brillouin line and the lowest folded line respectively. We have performed a detailed line shape analysis in order to extract in a systematic way the positions and the widths of the different lines in the spectra, after having removed the background due to the excitonic luminescence. Thanks to the coexistence of the laser line and the Brillouin lines in the same spectra, a very high accuracy has been obtained for the Brillouin shift, the distance between the inelastic lines and the elastic one.

We have deduced very systematic variations as a function of the incident energy, with a very high signal to noise ratio. As shown in Fig. 3, these variations exhibit systematic trends when the different lines and the Stokes and Anti-Stokes components for each one are compared. The main feature is an oscillation of the shift, with typical amplitude of 0.2 cm^{-1} , and a peak in the width close to the exciton resonance. Outside the resonance the linewidth is dominated by the resolution of the spectrometer amounting to 0.2 cm^{-1} while its peak value is of the order of 0.5 cm^{-1} . One should note that the intrinsic acoustic phonon lifetime is extremely long and has negligible influence on the linewidth. In the case of the folded modes, two lineshift oscillations are observed both in the

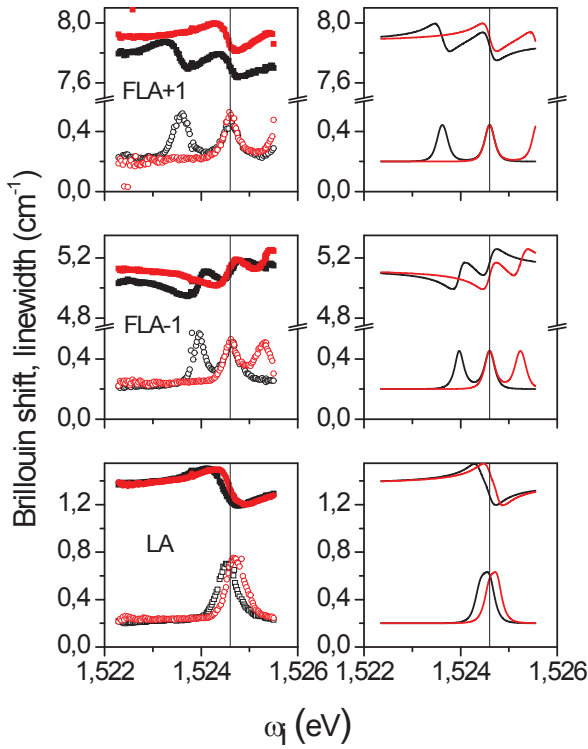


FIG. 3: Left panels: Variation with the incident energy of the Brillouin shift (full squares) and the Brillouin line width (open circles) deduced from the experimental spectra: LA phonon (lower panel), folded phonons FLA-1 (middle panel) and FLA+1 (upper panel). In each panel, the data are represented for the Stokes (red labels) and the Anti-Stokes (black labels) lines. Right panels: calculated values of the same quantities as in the left panel according to the model described in the text. The vertical line in all panels indicates the fitted exciton energy (incoming resonance).

Stokes and the Anti-Stokes components. One of these oscillations is common to both components while the second one appears at higher (resp. lower) incident energy for the Stokes (resp. Anti-Stokes) component. We attribute these two structures to the incoming and outgoing resonances. At the incoming resonance, when the laser energy coincides with the exciton, one expects a common feature in all the experimental traces, which is the case in our data for 1.5246 eV. The outgoing resonance is shifted from the incoming one by the Brillouin shift, positive or negative whenever the Stokes or the Anti-Stokes component is considered. This is verified in the experimental data. In the case of the LA line, the Brillouin shift is too small to allow the separation of the two resonance channels and a single resonance is observed at a slightly different position for Stokes and the Anti-Stokes components. The amplitude of the oscillation and the linewidth peak value are larger than for the folded lines because they

reflect the combination of two resonances. To summarize, the excitonic picture as schematically represented in Fig.1 gives an excellent qualitative description of the experimental features. In particular, it reproduces the observation of three identical oscillations in the energy shift and three identical peaks in the line broadening, one being common to the Stokes and the Anti-Stokes components and the other ones symmetrically shifted towards higher or lower energy in ST and AS respectively. We will now show that a theoretical model based on the discrete damped polariton dispersion allows to quantitatively well fit the main experimental features based on the expected background dielectric constant and exciton energy and provides a unique access to the exciton homogeneous broadening.

Based on the mechanisms presented in Fig. 1, it is straightforward to calculate the Brillouin shifts which fulfill both the conservation of the energy and the wavevector in a scattering event with a LA or a FLA line, both in the Stokes and Anti-Stokes channels. The result of this calculation is shown in the right panels of Fig.3, with the same colors as used in the left panel for experimental results. The full width at half maximum is obtained from the expression $2v(Im(k_i) + Im(k_s))$ in which k_i and k_s are the wavevectors of the incident and scattered polaritons. The experimental resolution (0.2 cm^{-1}) is taken account by a convolution formula strictly valid for Gaussian profiles only. We have taken the parameters given for bulk GaAs in Ref.[1] : a background dielectric constant equal to 12.55 and an exciton cross section corresponding to longitudinal transverse splitting of 0.086 meV. The exciton energy has been fitted to 1.5246 eV and the lifetime parameter γ to $0.30 \pm 0.03 \text{ meV}$. With these parameters, the width and the amplitude of the structures in the energy and linewidth resonance profiles are very well reproduced. The peak linewidth is slightly underestimated by an amount within the uncertainty in the lineshape fitting and the simplified treatment of the instrumental broadening. The bulk β value allows a good description of the experimental results as the variation due to confinement is expected to be small for such a wide quantum well [9]. Some discrepancy remains in the background variations of the line energies, negligible for the Brillouin line and of increasing magnitude with the energy shift of the line. This is likely to be due to additional contributions to the background dielectric constant, f. i. due to higher energy band to band transitions, not included in the model.

We would like to point out finally that the question of light scattering intensity resonances in the polariton frame has been much debated [2] and the conclusion remained unclear whether including the polariton picture significantly changes the resonance curves in bulk material, as compared to the exciton one. The analysis of our intensity measurements is a topic in itself and we leave it for a forthcoming publication together with additional results on resonances close to the E2HH2 and E3HH3

excitons.

In conclusion, we have observed very clear evidences of the polaritonic coupling in light scattering by acoustic and folded acoustic phonons in a multi-quantum-well when the polariton damping is larger than the longitudinal transverse splitting but significantly smaller than the Rabi splitting. Thanks to the combination of a specifically designed high resolution set up, a multi-quantum well with much thicker layers than in previous studies and an optimization of the experimental temperature, exceptionally clear variations of both position and width of Brillouin lines have been demonstrated for the first time when the photon energies satisfies either the incoming or the outgoing resonance. Additional novel results are the observation of similar consistent effects on acoustic folded lines. A quantitative description is obtained in a damped polariton picture provided a broadening of 0.3 meV is included, a value significantly smaller than the exciton linewidth measured in photoluminescence excitation. This suggests that light scattering gives access to new information on the damping mechanisms in multi-quantum wells, possibly excluding the inhomogeneous broadening from well to well owing to the delocalized nature of the probing acoustic phonons.

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